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Comment on "Direct Observation of Crack-Tip Geometry of SiO_2 Glass by High-Resolution Electron Microscopy"

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IN A RECENT communication Bando *et al.*[1] claim to observe the intrinsic tip structure of stationary cracks in fused silica glass by high-resolution transmission electron microscopy (TEM). They produced foils by etching in HF to a thickness 20 to 40 nm and introduced the cracks by damaging these foils with a needle. The observed crack-tip contours appear to be somewhat rounded, with a radius of curvature of $\simeq$1.5 nm in the freshly damaged state. This result was put forward as evidence in support of theoretical estimates of crack-tip radii by Pavelchek and Doremus.[2] After the cracks were aged for 7 d in saturated silicic acid solution at 90°C, the measured radius was $\simeq$5 nm. The increase in radius was taken to indicate that some chemical blunting must have occurred, in accordance with a model of dissolution and precipitation proposed by Ito and Tomozawa.[3]

The purpose of the present comment is to question the interpretations of the microscopic observations of Bando *et al.*[1] in terms of well-defined tip radii. At issue here is the fundamental nature of the crack configuration in the critical region where material separation occurs.[4] In this context it may be noted that the authors of an analogous TEM study of cracks in several brittle *crystalline* materials[5] came to an altogether different conclusion, namely that the crack tips are atomically sharp (i.e. consistent with the picture of fracture by sequential bond scission). There appears to be no reason to suggest that glass, merely because it is noncrystalline, should behave differently. In another study, using controlled indentations in glass,[6] it was demonstrated that strength increases resulting from prolonged exposure of the cracks to aqueous environments are unequivocally associated with a progressive relaxation of residual contact stresses, i.e. the aging phenomena do *not* constitute evidence for blunting. The clear implication of these other studies is that the observations in Ref. 1 may not accurately reflect the true structure of brittle cracks.

However, the most compelling argument against the blunt-crack interpretations comes from scrutiny of the micrographs shown in Ref. 1. Special reference is made to Fig. 2(*A*) in that communication, which is a side-on view of a newly formed crack in the glass foil. From this micrograph, wall-wall displacements, $2y$, can be measured directly as a function of the distance, x, behind the apparent crack tip, as indicated in our Fig. 1(*A*). Now since this particular crack is purported to be representative of the pristine state (i.e. the walls have supposedly not undergone any post-formation structural change), the displacement fields given by conventional fracture mechanics solutions should provide an accurate quantitative description of the functional relation $y(x)$, at least down to the "tip" region, where x and y become comparable (i.e. where the strains lie in the nonlinear region). For plane stress conditions (appropriate to thin foils) this relation is[7]

$$y^2=(8/\pi)(K/E)^2x \qquad (1)$$

where E is Young's modulus and K the stress intensity factor responsible for the residual crack opening. A plot of y^2 vs x using direct measurements from Fig. 2(*A*) in Ref. 1 is indeed linear, as shown in our Fig. 1(*B*). From the slope of this line, $(8/\pi)(K/E)^2=3.7\pm0.3$ nm, in conjunction with the value $E=72$ GPa for fused silica,[8] we evaluate $K=2.7\pm0.2$ MPa·m$^{1/2}$. This value is 3 to 4 times the limiting stress intensity factor (i.e. the toughness) for silica glass, $K_c=0.8\pm0.4$ MPa·m$^{1/2}$,[8] below which cracks can remain stationary. Thus, if the fissures shown in Ref. 1 were to be at all representative of true cracks in glass, they would lie well in the region of fast propagation. It must be concluded that the observations in Ref. 1 are associated with artifacts in the experimental method.

The source of such artifacts cannot be established without a more detailed investigation into the exact procedures used to obtain the TEM crack images, but there are possibilities that deserve closer attention. First, the limit of spatial resolution in the high-magnification micrograph shown as Fig. 1(*B*) in Ref. 1 (as apparent from the Fresnel fringes) is $\simeq$1 nm, i.e. of the same order as the estimate of the intrinsic tip radius. Second, contamination of newly created surfaces, either by the specimen preparation technique or during the electron microscopic examination itself, is always difficult to avoid.[9] Such contamination could conceivably fill up the narrowest interfacial regions of the crack (much as envisaged by Bando *et al.*[1] in their explanation of the aged cracks in their Fig. 2(*B*)) and thereby create a false tip. (There is some suggestion of this in the micrographs in Ref. 1, even those for the "newly formed" cracks, in the form of distinctive bands of contrast adjacent to the walls and ahead of the apparent tip.)

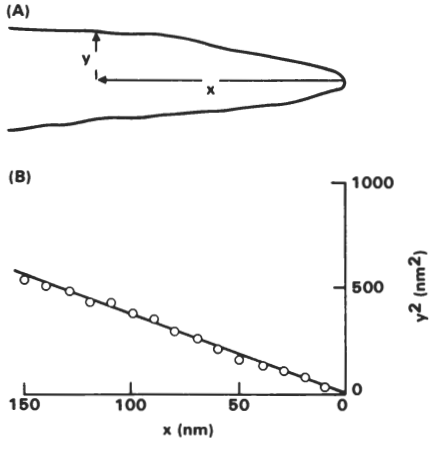

Fig. 1. (*A*) Profile of newly formed crack in high-silica glass observed by transmission electron microscopy (traced directly from Fig. 2(*A*) in Ref. 1). (*B*) Plot of crack wall displacement, y, as function of square root of distance, x, behind apparent tip of same crack.

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